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EXPERIMENTAL STUDIES OF THE SATURATION LEVEL OF METHANE HYDRATE IN THE EASTERN NANKAI TROUGH SEDIMENTS

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ABSTRACT

The pore saturation of natural gas hydrate in sediments is a key parameter for estimating hydrate resources in a reservoir. For a better understanding of gas hydrate distribution, the experimental study of the pore saturation of methane hydrate in sediments from a hydrate reservoir in the Eastern Nankai Trough have been carried out. In total, eleven samples, comprising sand, silty sand, silt, and representative of the main sediment types identified in the Eastern Nankai trough, were tested. The results obtained clearly indicate a particle size and clay content dependent trend: almost 100% of pores were saturated with methane hydrate in sand when little silt and clay were present, decreasing to ~ 13% in silty sand (sand 54%, silt 41% and clay 5%), and ~ 4% in clayey silt. These results are generally consistent with NMR logging results for high-saturation samples, but somewhat different for samples with medium or low saturation levels.

Keywords: Methane hydrate, saturation, sediment types, particle size, and specific surface area

1. INTRODUCTION

The accurate estimation of natural gas hydrate resources is one of the most important issues in assessing the energy potential of natural gas hydrate, which relies largely on the precision of the data on the pore saturation of natural gas hydrate in sediments of a reservoir [1]. Because some hydrate dissociation always happens during core recovery, even the saturation data obtained on the recovered hydrate-bearing sediments are uncertain. Currently, saturation data obtained from geophysical investigations are widely applied to methane hydrate resource estimates, but they are also uncertain because the explanation of geophysical data is carried out with uncalibrated models. In such a situation an experimental study of hydrate saturation in sediments from a hydrate reservoir is with great significance.

In natural environments, the formation of gas hydrate inevitably will be subject to the influence of sedimentary characteristics, so the modes of gas hydrate formation and occurrence may well be different in sediments with various particle sizes and mineral compositions. The elucidation of this issue, how sediments affect the formation and occurrence of gas hydrate, will help in more efficient gas hydrate exploration, accurate estimation of gas hydrate resources and the design of gas hydrate production methods.

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Intensive investigations have been implemented to study the distribution of hydrate and to estimate hydrate resources in the Eastern Nankai Trough [1]. Although hydrate saturation in the reservoir have been estimated from regional seismic and well-logging results, calibration with results obtained under controlled conditions is needed. In this paper we present the experimental results of hydrate saturation in sediments recovered from the Eastern Nankai Trough gas hydrate region for the calibration of well-logging results.

2. METHODOLOGY

Most natural minerals are kinetically stable even under conditions far away from their usual regime of thermodynamic stability, for example quartz and garnet. However, gas hydrate is rather different from these, dissociating into water and gas as soon as it is taken outside of its stability region. Unfortunately, in most cases gas hydrate recovery processes take hydrate out of its stable regime, consequently only rarely can natural hydrate samples be recovered intact. Hydrate dissociation not only results in the disappearance of the natural gas hydrate sample but also the texture of gas hydrate bearing sediments is reformed by the release of gas and water. This makes it difficult to study the characteristics of natural gas hydrate by direct analysis of natural samples. As a result, laboratory-based experimental studies, to study a specific property of gas hydrate under controlled conditions, have a special significance for gas hydrate research.

According to gas analyses results [2], the gas hydrates recovered in the Eastern Nankai Trough were structure I (sI) methane hydrate, so the gas employed for this research was methane gas. The sediment specimens tested were from the Eastern Nankai Trough, recovered during the 2004 gas hydrate drilling campaign in the Eastern Nankai Trough. They were stored in a frozen state after being subsampled at the drilling site. The sediment core was trimmed into a column with a diameter of one inch. To avoid the possible movement of sediment particles by hydrate formation, the frozen sediment core was bound tightly with a fiberglass screen.

The experiments were carried out by reacting the natural sediments with methane gas in a pressure vessel. The sediments were thawed in a portable fridge before they were sealed into the pressure vessel. After the pressure vessel was evacuated for about 10 minutes, methane gas was charged into the vessel up to 12 MPa. Then the pressure vessel was moved into a room in which the temperature was kept at a constant 3 °C.

When the pressure did not change for one week, it was concluded that the reaction was complete. To avoid any dissociation of gas hydrate during sample recovery, the pressure cell was put in a liquid nitrogen box to cool it down. After the vessel was cold, the samples prepared were recovered and stored in liquid nitrogen.

The hydrate-containing sediments prepared were analyzed for hydrate saturation estimates as to the method described in [3]. Due to gravity drainage, some water may have drained away from the top layer, and extra water may be present between the sediment and the wall of the container. As a result, only the data obtained from the central portion of the sediment core were used in this paper. The amount of gas hydrate was estimated from the amount of gas released from a sample with a hydration number of 6.0 [4], while the volume of gas hydrate was calculated for a density of 0.92 g/cm^3.

As described in Table 1, in total 11 specimens were tested. The specific particle size distribution in each sample was measured by a laser particle size analyzer.

Table 1 Particle size distribution in the sediments tested.

Specimen No.	Sediment type*	Sand (%)	Silt (%)	Clay (%)
1	Clayey silt	0.81	67.80	31.39
2	Silty sand	53.58	41.38	5.04
3	Silty sand	60.46	33.96	5.59
4	Silty sand	67.32	28.22	4.46
5	Sand	75.36	22.09	2.54
6	Sand	84.80	12.50	2.70
7	Sand	84.86	12.92	2.22
8	Sand	89.52	8.71	1.77
9	Sand	89.74	6.89	3.37
10	Sand	91.68	6.67	1.65
11	Sand	93.09	5.18	1.74

*As referred to the criterion of Shepard, 1954.

3. RESULTS AND DISCUSSION

The results obtained are summarized in Table 2. For comparison, the pore saturation data, obtained by the DMR (Density-magnetic Resonance) method through NMR and density logging [1] from the sediments at the similar depth and with similar lithology compared to the sediments studied, are also listed in Table 2. It can be seen that the pore saturation of methane hydrate in the sediments studied are in a wide range from 4.1% in clayey silt to almost 100% in sand. It looks like the experimental results are generally consistent with the logging results, although exceptions have been found for the two fine sediments (sample #1, clayey silt, and sample #2, silty sand) and one sand sample (#11).

Table 2 The comparison of the results for the hydrate pore saturation in synthesized and natural samples

	Hydrate pore saturation	
Specimen No.	Experimental (%)	DMR Method (%)*
1	4.1	31.6
2	12.7	61.0
3	39.8	43.9
4	64.7	54.4
5	33.3	31.6
6	100.0	80
7	34.7	46.7
8	39.6	42.8
9	50.4	57.1
10	45.3	55.2
11	25.0	50.6

Direct measurements of hydrate saturation in sediments from Mallik and Cascadia indicate that hydrate saturation is very low in fine sediments, only several percent of the pore space, agreeing well with the experimental results of this research. Therefore it seems that the DMR method overestimates hydrate saturation in fine sediments and appears to be limited by the method. Due to the short transverse nuclear magnetic relaxation times (T_2) of protons in the solid hydrate lattice of gas hydrate (<< 0.2 ms) this signal will not be detectable by NMR. As a result the NMR porosity is smaller than the γ-γ density porosity. Based on this phenomenon the DMR method estimates hydrate saturation from the difference between γ-γ density porosity and apparent NMR porosity [5]. However in sediment, pore water can be bound to the surface of sediment particles, consequently some of this may not be detectable by NMR either and will become invisible. This will result in an overestimate of hydrate saturation. In coarse sediments, the percentage of bound water is low as compared to bulk water, so the overestimate will not be that serious. But in fine sediments the overestimate may be pronounced due to the high surface area and the relatively high percentage of bound water.

As a check for the condition of sample #11, the water in it was lost during transportation and storage. It is the reason for a lower saturation than for that in other sand samples.

Table 3 The hydrate saturations and average particle sizes for the sediments studied

Specimen No.	Hydrate saturation (%)	Particle size (μm)
1	4.1	12.99
2	12.7	73.78
11	25.0	202.3
5	33.3	115.2
7	34.7	116.9
8	39.6	129.7
3	39.8	107.2
10	45.3	145.6
9	50.8	104.7
4	64.7	160
6	100.0	197.7

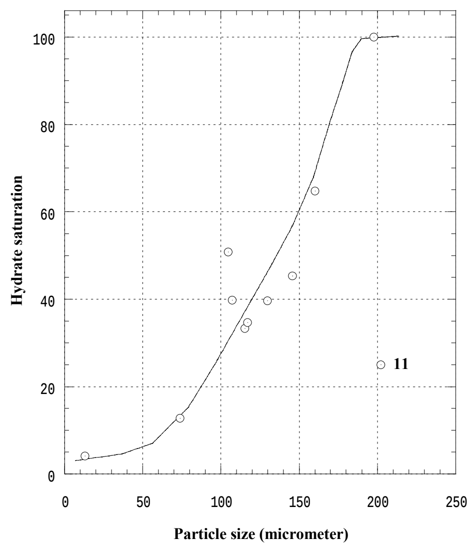

Figure 1 The relationship of pore saturation of methane hydrate and average particle size in sediments studied

Table 3 lists the hydrate saturation and average particle size of sediments, and they have been plotted in Figure 1. An obvious particle size-dependent trend can be recognized: low hydrate saturation when particle sizes are smaller than 50 μm; increasing dramatically from 50 μm to about 200 μm, and almost reaching a maximum saturation when the particle size > 200 μm. This phenomenon is exactly the same as that observed with silica sands [6], and which has been explained as the effect of the specific surface area of sediments. With the increase in specific surface area more water is bound to the surface of sediment particles, thereby less water is available for hydrate formation and lower hydrate saturation will be the result.

4. CONCLUSIONS

In summary, our investigations on the hydrate saturation in the natural sediments from the Eastern Nankai Trough have revealed a close relationship between the hydrate saturation with sediment particle size. These results are generally consistent with logging results for high hydrate saturation samples but somewhat different for samples with medium or low saturation levels.

However the sediments studied so far have still been limited to just a few types, so a systematic investigation on how sediment type, primarily particle size, controls the saturation of gas hydrate is necessary. In future research, we will try to 1) study the mineralogical influence on hydrate stability in the systems of both pure water and aqueous solutions, and 2) investigate hydrate saturation in the sediments with various particle sizes and various mineral compositions, completing at least one point for each sediment category as described by Shepard (1954).

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